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BY:

Gene Conti

Date:

June 15, 2004

MAIL STOP RCE

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of:
Atsushi Ueda et al.

Conf. No.: 7373

: Group Art Unit: 1745

Appln. No.: 10/058,707

: Examiner: Raymond Alejandro

Filing Date: January 28, 2002

: Attorney Docket No.: 10059-404US
(P27007-01)

Title: Non-aqueous Electrolyte Secondary Battery

DECLARATION OF ATSUSHI UEDA UNDER 37 C.F.R. § 1.132

I, Atsushi Ueda, declare and state as follows:

1. I am a co-inventor of the invention described and claimed in the above-identified patent application.

2. I received a Master's Degree in chemistry of molecules from the Engineering Department of the graduate school of Osaka University in March 1998 with.

3. Since April 1998 I have been employed as an engineer in the Lithium Secondary Battery Business Unit of Matsushita Battery Industrial Co., Ltd., a subsidiary of Matsushita Electric Industrial Co., Ltd., where I have been engaged in the research and development of lithium ion secondary batteries.

4. I am familiar with the above-referenced application, and in particular with the Office Action dated February 19, 2004 (Paper No. 20040107). I am submitting the present Declaration to overcome any § 103(a) rejections of claims 14 and 16 by demonstrating the criticality of the molar ratio between LiPF_6 and LiBF_4 in the solute of a non-aqueous electrolyte

secondary battery according to the invention, which would not be expected based on the prior art.

The Examiner's Position and the Purpose of the Declaration

5. The Examiner has taken the position that the closest prior art to the invention includes U.S. Patent Application Publication No. 2002/0064712 of Sekino et al. ("Sekino"), U.S. Patent Application Publication No. 2003/0118913 of Takami et al. ("Takami"), U.S. Patent No. 6,315,918 of Mita et al. ("Mita"), EP 0 796 510 ("EP '510") and U.S. Patent Application Publication No. 2002/0001756 of Hamamoto et al. ("Hamamoto").

6. In the Office Action dated February 19, 2004, the Examiner rejected claims 14-16 under 35 U.S.C. § 102(e) as being anticipated by Sekino and under 35 U.S.C. § 103(a) as being obvious over various combinations of Takami, Mita, EP '510 and Hamamoto. The Examiner argued that all of the claimed elements are taught or suggested by the cited references, including the employment of lithium salts, such as LiPF_6 and LiBF_4 , in the solute, as recited in claims 14 and 16.

7. The purpose of this Declaration is to demonstrate that the claimed molar ratio of LiPF_6 to LiBF_4 , 1:9 to 9:1, which is not taught by the cited prior art, is critical to these embodiments of the present invention and would not be expected. Such a demonstration will thus overcome the rejections of claims 14 and 16.

Background and Purpose of the Invention

8. The non-aqueous electrolyte secondary batteries according to the present invention were developed to overcome problems known in the art, such as deterioration of charge and discharge characteristics. The batteries according to the invention have excellent charge and discharge characteristics, particularly at low temperature, and show satisfactory charge and discharge characteristics, even after being exposed to high temperatures over a period of time.

9. According to the presently claimed invention of claims 14 and 16, non-aqueous electrolyte secondary batteries which exemplify these properties are realized by using a non-aqueous solvent containing: (A) a cyclic carboxylic acid ester; (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond; and (C) a cyclic carbonic acid ester having no carbon-carbon unsaturated bond. In one embodiment (claim 14), component (B) comprises

vinylethylene carbonate (VEC). As recited in claims 14 and 16, in a preferred embodiment, the solute dissolved in the non-aqueous solvent contains both LiPF_6 and LiBF_4 in a molar ratio of 1:9 to 9:1. While LiPF_6 has excellent electrical characteristics, it is chemically unstable since it produces hydrofluoric acid. Cyclic carboxylic acid esters (A) are prone to decomposition by hydrofluoric acid, possibly deteriorating the cycle characteristics of the battery. Additionally, the use of LiBF_4 also deteriorates cycle characteristics of the battery because the degree of electrolytic dissociation of LiBF_4 is small or a film formed on the surface of the negative electrode comprising graphite is weak. However, as shown in Tables 10 and 11 of the present application, the use of both salts in combination (particularly in the claimed ratio, as will be demonstrated below), improves cycle characteristics of the batteries and decreases the amount of gas generated, and also results in favorable capacity maintenance rates and cycle life.

10. In order to demonstrate the effects of the molar ratio of LiPF_6 to LiBF_4 in the solute of the non-electrolyte secondary batteries, batteries were prepared in which the molar ratio of LiPF_6 to LiBF_4 were 0.95:0.05 and 0.05:0.95. The capacity maintenance rates, cycle lives, and amounts of gas generated after cycles were measured in order to assess the performance of these batteries.

Experimental Procedure

11. Batteries were produced using the procedure described in Example 5 of the above-identified application, except that the LiPF_6 : LiBF_4 ratios were (a) 0.95:0.05 and (b) 0.05:0.95. These batteries contained, as the non-aqueous solvent, γ -butyrolactone, vinylene carbonate, ethylene carbonate and diethyl carbonate in a 68:2:20:10 volume ratio.

12. As in Example 5, these batteries were subjected to the measurement of capacity maintenance rates after storage at high temperature, cycle lives, and amounts of gas generated after the cycles.

Results and Discussion

13. The Table below shows the results obtained for the capacity maintenance rates after storage, cycle lives, and amounts of gas generated after cycles for batteries (a) and (b), as well as for those depicted in Table 11 of the present application, which include batteries containing only LiPF_6 ("1F"), only LiBF_4 ("5F"), as well as LiPF_6 and LiBF_4 in ratios of 9:1 ("2F"), 1:1 ("3F"), and 1:9 ("4F").

14. As can be seen from comparing the data in the Table, when the relative amount of LiPF_6 is high (see batteries 2F and 3F, for example), the capacity maintenance rate after storage at high temperature is high and the amount of gas generated after the cycles is low. However, when the relative amount of LiPF_6 is greater than 0.9 M (ratio of $\text{LiPF}_6:\text{LiBF}_4$ greater than 9:1), as in batteries (a) and 1F, the cycle life is unfavorably reduced to 320 or 300, respectively.

| Battery | Mixing Amounts of $\text{LiPF}_6/\text{LiBF}_4$ (M/M) | Ratio of $\text{LiPF}_6/\text{LiBF}_4$ | Capacity Maintenance Rate After Storage (%) | Cycle Life | Gas Amount After Cycles (mL) |
|---------|---|---|--|------------|------------------------------------|
| 1F | 1/0 | 1/0 | 84 | 300 | 2.3 |
| (a) | 0.95/0.05 | 0.95/0.05 | 84 | 320 | 2.2 |
| 2F | 0.9/0.1 | 9/1 | 84 | 580 | 2.0 |
| 3F | 0.5/0.5 | 1/1 | 83 | 750 | 2.2 |
| 4F | 0.1/0.9 | 1/9 | 82 | 450 | 2.5 |
| (b) | 0.05/0.95 | 0.05/0.95 | 81 | 470 | 2.8 |
| 5F | 0/1 | 0/1 | 81 | 500 | 3.0 |

15. On the other hand, as the relative amount of LiBF_4 is increased, the capacity maintenance rate tends to decrease and the amount of gas generated after the cycles tends to increase. When the relative amount of LiBF_4 exceeds 0.9 M (ratio of $\text{LiPF}_6:\text{LiBF}_4$ less than 1:9), as in batteries (b) and 5F, the capacity maintenance rate becomes as small as 81% and the amount of gas generated after the cycles increases to as large as 2.8 or 3.0 mL, respectively.

16. It can thus be concluded from the above Table that when the $\text{LiPF}_6:\text{LiBF}_4$ molar ratio is maintained between 9:1 and 1:9, the resulting batteries exhibit excellent capacity maintenance rates and cycle lives, and small amounts of gas are generated after the cycles. However, when the molar ratio falls outside this range, inferior results are obtained.

17. Although these experiments were performed in the absence of VEC, Example 6 of the present application demonstrates the favorable effect of VEC in reducing the amount of gas generated. Therefore, it can be assumed that inclusion of LiPF_6 and LiBF_4 in a molar ratio of 9:1 to 1:9 in a battery which also includes VEC would also result in similar favorable results.

Conclusion

18. This Declaration demonstrates that by preparing non-aqueous electrolyte secondary batteries according to one embodiment of the invention as claimed in claim 16, in which the solute contains both LiPF_6 and LiBF_4 in a molar ratio of 9:1 to 1:9, favorable results

are obtained, particularly with regard to capacity maintenance rate, cycle life, and amount of gas generated after cycles. Similar results would be expected for a battery which contains VEC in the non-aqueous solvent, as claimed in claim 14. None of the prior art specifically teaches the use of both lithium salts in combination, and further does not teach the claimed molar ratio. Accordingly, the criticality of the claimed ratio would not have been expected based on the cited prior art or to one having ordinary skill in the art at the time of the invention based on the prior art of record.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that those statements were made with the knowledge that willful false statements the like so made are punishable by fine or imprisonment, or both, under Section 1003 of Title 18 of the United States Code; and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: June, 14, 2004

Atsushi Ueda
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